EVALUATION OF MICROBIALLY INFLUENCED DEGRADATION AS A METHOD FOR THE DECONTAMINATION OF RADIOACTIVELY CONTAMINATED CONCRETE

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ABSTRACT

Because there are literally square kilometers of radioactively contaminated concrete surfaces within the U.S. Department of Energy (DOE) complex, the task (both scope and cost) of decontamination is staggering. Complex-wide cleanup using conventional methodology does not appear to be feasible for every facility because of prioritization, cost, and manual effort required. We are investigating the feasibility of using microbially influenced degradation (MID) of concrete as a unique, innovative approach for the decontamination of concrete. Currently, work is being conducted to determine the practicality and cost effectiveness of using this environmentally acceptable method for decontamination of large surface concrete structures. Under laboratory conditions, the biodecontamination process has successfully been used to remove 2 mm of the surface of concrete slabs. Subsequently, initial field application data from an ongoing pilot-scale demonstration have shown that an average of 2 mm of surface can be removed from meter-square areas of contaminated concrete. The cost for the process has been estimated as $1.29/m2. Methodologies for field application of the process are being developed and will be tested. This paper provides information on the MID process, laboratory evaluation of its use for decontamination, and results from the pilot field application.

INTRODUCTION

Uncoated concrete has been used for the construction of ponds, canals, sumps, and other structures within operating nuclear facilities. Many of these concrete structures have become contaminated over time with various radionuclides. The most frequently occurring radiological contaminants appear to be Cs-137, U-238, and Co-60, followed closely by Sr-90 and tritium. Typically, this contamination is securely Flxed on the surface or within the first 1 or 2 mm.
The total area of contaminated concrete within the DOE complex is estimated to be in the range of 73 km2. The volume of contaminated concrete is estimated at 1.9 x 105 m3. Because of the immensity of contaminated concrete surfaces within the DOE complex, the task (both scope and cost) of decontamination is staggering. Concrete decontamination needs have been identified as (1) reduction of secondary waste, (2) cost- and schedule-effective technologies, and (3) innovative technologies for floor and wall decontamination Based on the above information, complex-wide cleanup using conventional methodology does not appear to be feasible for every facility because of prioritization, cost, and the manual effort required.
A practical, cost-effective, environmentally acceptable method for decontamination of large-surface concrete structures could be accomplished through the use of a naturally occurring microbiological process that destroys concrete integrity. Known as microbially influenced degradation (MID) of concrete,23 the process occurs when microorganisms present in the environment produce minerals or organic acids that dissolve or disintegrate the cement matrix. The literature indicates that MID-promoting microorganisms are ubiquitous in the environment

and that their mechanisms of attack are consistent with those that have been associated with chemical attack. It seems reasonable that a method could be developed using the same process, under controlled conditions, to decontaminate certain concrete structures.
Data from previous work on the development of a biodegradation test for cement-solidifled waste forms2 demonstrated that the most active of the MID microorganisms were sulfur-oxidizing bacteria, which were therefore selected for development of the biodecontamination process. The concept of biodecontamination is seen in Figure 1. This paper discusses laboratory evaluation and field prototype testing of a biodecontamination process.

MATERIAL AND METHODS

Thus far in the development cycle, two laboratory studies and a field prototype study have been conducted. The methods, application, and effects of accelerated MID testing on a variety of cement formulations were developed previously.3 In the first laboratory study, chambers were designed and constructed to expose concrete specimens 10 x 10 cm (100 cm of exposed Ace) to the effects of accelerated MID. The concrete specimens were made using a commercial mix of portland type II cement and 6.3 mm aggregate. After curing the specimens for 72 hours, the surface was sprayed with 5 mL of an aqueous solution containing 100 ppm cobalt [as Co(NO3)2] for a total application of 500 lag cobalt per surface. This was done so the specimens could serve as surrogates for concrete contaminated with the radionuclide Co-60.
Two of the Co-60 "contaminated" concrete specimens were used as a treatment and control in this initial laboratory study. Treatment consisted of exposing the specimen to solutions delivered in the form of an intermittent, fme spray of *Thiobacillus thiooxidans* lixiviant. The control was sprayed with a sterile media solution. Specimens were placed in the exposure chambers at a 45-degree angle to prevent pooling and to promote liquid runoff into a collection reservoir. The collection reservoir was emptied daily, and the quantity of liquid was recorded, with 100 mL saved for later analysis. The test cells are seen in Figure 2.
The concrete used in the second laboratory study was obtained from a concrete foundation constructed in the mid-1950s. The area being evaluated was the original weathered surface, which had a pH near 10. To facilitate a dimensional determination of surface loss due to MID, four

Concentration of
Concrete Surface

Figure 1. Diagram detailing the conceptualized activity of MID bacteria and how their activity can be used to facilitate the biodecontamination of concrete surfaces

Figure 2. Setup of the laboratory-scale H2S chamber for evaluating the ability of MID to remove surface layers of environmentally aged concrete (95-633-1-4).

cores 2 cm in diameter by 4 cm long were cut out of four quadrants of the specimens. These cores were raised 5 mm above the surface and fixed in place with cement mortar. Epoxy resin used to seal concrete surfaces was applied to the protruding surface of one of the four cores. Since many contaminated concrete surfaces are coated with epoxy resin, this treatment was intended to determine what effect MID would have on the coating.
The study was initiated by inoculating the concrete surface with a T. *thisoxidans* monoculture. The chamber was then flooded with a continuous supply of compressed air amended with a

sufficient mixture of 100 ppm H2S (vh in N2) to establish a constant internal concentration of 10 ppm H2S (v/ v). Humidity in the chamber was maintained at 100to.
After successful operation of the laboratory-scale H2S chamber for a period of 3 months, a field prototype based on its design was fabricated. The location for demonstration of the field prototype was at the Idaho National Engineering Laboratory's Experimental Breeder Reactor I (EBR-I) reactor building. EBR-I was the first power reactor to produce sufficient electricity to light a city. The facility became operational in 1951, and decommissioning was completed in 1964. After a radiological survey, three areas were selected that had fixed contamination ranging from several hundred to a few thousand counts per minute as determined by a hand-held survey meter.
The Weld demonstration used three larger versions of the air-tight, wall-mounted chamber developed and tested in the laboratory. Chamber sizes were selected to cover areas of contaminated concrete. Two had dimensions of 0.5 m x 0.5 m x 0.05 m, while the third was 1.0 m x 1.0 m x 0.05 m. An attached chamber is seen in Figure 3.

Figure 3. Test setup for evaluation of MID-promoted decontamination of concrete surfaces at the EBR-I field facility (96-114-1-28).

For purposes of designation, the chambers were numbered 1 through 3. Chambers 1 and 3 were designed to evaluate two different sulfur sources (potassium tetrathionate [K2S406] or hydrogen sulfide [H2S]), while Chamber 2 was used as an abiotic control. The surfaces encompassed by Chambers 1 and 3 were inoculated with a monoculture of T. *thiooxidans* using the method developed for the laboratory chamber. Valves attached to chamber access ports were used to facilitate sampling of the interior atmosphere. Once per week, sterile cotton swabs were used to take swipes of the concrete surface of each chamber. These samples were used to determine the numbers of viable bacteria growing on the wall surfaces.

RESULTS

Sprav Chambers

Chemical analysis showed that over the 10 weeks of this study, nearly 1 g (920 mg) of calcium was removed from the specimen treated with thiobacilli. The rate of loss was nearly linear with time, suggesting that calcium loss would have continued at the same rate had the study been extended. In addition, data showed that at least 100% of the applied cobalt was removed, thus resulting in an almost 1005ro NIID-induced decontamination of the test specimen (Figure 4).

Figure 4. Results of an accelerated "spray concrete surfaces was examined by probing with chamber" test showing the cumulative the blade of a spatula. After 6 months of removal, over time, of a cobalt contaminant operation, it was determined by this method that from the surface of concrete.

In General, it was found that there was an overall heterogeneity of surface softness. A 105-cm area was scraped with a spatula blade to remove loosened surface material (between 1 and 3 mm deep). MID activity was also evident on the protruding 2-cm-diameter cores, one of which had the epoxy coating. It was found that 0.05 g of material could be removed from the unprotected core, while 0.29 g of solids could be removed from coated core.

These data confirmed that MID could be initiated on the concrete surface and that the result of this activity was the production of an easily removable, soft, friable surface. Importantly, it was also shown that epoxy coatings can be compromised by MID in a relatively short period of time and that such a coating could, in reality, promote intense MID activity.

Field Prototype Chambers

Chambers 1 and 2 were set up, tested, and in operation 5 weeks before Chamber 3 became operational. Routine sampling of chamber operation has been ongoing since the initiation of the study. Sufficient numerical data on bacterial numbers and H2S concentrations have been collected to begin the development of two-dimensional representations of these data. It was found that there was a uniform mixing of H2S in the chamber with average zonal concentrations between 55 and 56 ppm (vh). Others4 have shown that this H2S concentration is in excess of that required to maintain high numbers of sulfur-oxidizing bacteria on concrete surfaces.
Physical observation of the decontamination process was made after 5 months for the inoculated H2S chamber (Chamber 3) and 7 months for Chamber 1 supplied with K2S406 (sampling of the chambers occurred on the same date, but different periods of operation were due to staggered start dates). At the time of sampling, the surface of the contaminated concrete could be observed and samples were retrieved (Figure 5). Probing at various locations on the surfaces showed that softening was occurring. The process of MID had progressed to a point that it was possible to remove loosened, friable concrete from the surface. As much as 1 to 4 mm of the sampled surfaces could be removed. The mass of material obtained from this process was as much as ~1 g/cm2. Of more importance, however, was that there was a significant reduction in the amount of fixed con-tamination on the treated surfaces. Surface readings Chamber 3 were reduced In Chamber 1, the initial contaminant level was reduced from 1,500 cpm to background after surface removal. These findings greatly exceeded our initial expectations. Based on other data, it was assumed that at least 12 months of operation would be required to allow for this significant amount of MID occurrence us, and decontamination. Operational conditions of the chambers were resumed after this brief examination. Further examination will occur after 12 months of operation.

CONCLUSIONS

Prototype applications substantiate that MID can be initiated and managed over a large surface area of concrete walls. In addition, it was shown that managed MID could be used to promote the removal of 2 to 4 mm of the concrete surface. These data serve as a basis to support the concept of biodecontamination.

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